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DERWENT-ACC-NO: 1993-365380
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TITLE: Thermoplastic resin compsn. with good impact resistance and improved heat-sealing property - contains cyclic olefin! copolymer obtd. by copolymerising cyclic olefin and alpha-olefin, useful for car components etc.

PRIORITY-DATA: 1992JP-0098653 (March 25, 1992)

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PATENT-FAMILY:

	PUB-NO	PUB-DATE	LANGUAGE	PAGES	MAIN-IPC
☐	JP 05271484 A	October 19, 1993		012	C08L023/02
☐	JP 3221721 B2	October 22, 2001		014	C08L023/08

INT-CL (IPC): C08L 23/02; C08L 23/08; C08L 23/18; C08L 45/00; C08L 101/00

ABSTRACTED-PUB-NO: JP 05271484A
BASIC-ABSTRACT:

Compsn. consists of 2-98 wt.% of (a) cyclic olefin type copolymer with glass transition temp. (Tg) up to 30 deg.C obtd. by copolymerising cyclic olefin with alpha-olefin and 98-2 wt.% of (b) thermoplastic resin.

Typically, cyclic olefin contains repeat unit of formula (Y) where Rb, Rm is H, 1-20C hydrocarbon gp. or halogen, oxygen or nitrogen contg. substit.; n is above 0; Rj and Rk or Rl and Rm may form a ring. Example of cyclic olefin is norbornene, 5-methyl norbornene, 5-ethyl norbornene, 5-propyl norbornene, 5,6-dimethyl norbornene, 1-methyl norbornene, 7-methyl norbornene, 5,5,6-trimethyl norbornene, 5-5-phenyl norbornene, 5-benzyl norbornene, etc..

USE/ADVANTAGE - The compsn. can be used for car parts, domestic appliances, films, etc. effectively due to good impact resistance and heat sealing property.

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2.**** shows the word which can not be translated.
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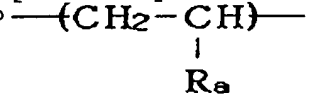
DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application] This invention has the outstanding shock resistance and the outstanding heat-sealing property, and relates to the thermoplastics constituent which can be effectively used in various fields, such as an automobile, household electric appliances, and a film.
[0002]
[Description of the Prior Art] Generally, since thermoplastics is excellent in a moldability compared with other materials and has sufficient rigidity, it is widely used as a molding material of a product with various electric products including household articles, industrial products, etc. However, although thermoplastics has the outstanding property mentioned above, it cannot be said to be enough [a shock-proof field]. On the other hand, the constituent which consists of ethylene and an annular olefin random copolymer, and ethylene and an alpha olefin random copolymer is indicated by JP,1-96234,A as a resin constituent which improved shock resistance. Here, glass transition temperature (Tg) is 50-250 degrees C, and the above-mentioned ethylene and annular olefin random copolymer have a problem in shock resistance. And in invention of JP,1-96234,A, in order to improve the shock resistance of this ethylene and annular olefin random copolymer, ethylene and an alpha olefin random copolymer are blended. Therefore, generally this invention does not improve the shock resistance of thermoplastics. This invention was made in view of the above-mentioned situation, and aims at offering the resin constituent which has improved the shock resistance of thermoplastics.
[0003]
[Means for Solving the Problem and its Function] As a result of inquiring wholeheartedly, when the annular olefin system copolymer of the specific structure of having a specific glass transition temperature (Tg) is blended with thermoplastics, this invention persons do the knowledge of the shock resistance of thermoplastics, and a heat-sealing property improving further, and came to make this invention. That is, the resin constituent of this invention is considered as 98 % of the weight and the configuration which consists of an annular olefin system copolymer 2 whose glass transition temperature (Tg) it is the copolymer obtained by copolymerizing (a) annular olefin and an alpha olefin, and is 30 degrees C or less - 98 - 2 % of the weight of (b) thermoplastics.
[0004] In addition, the constituent (heat-resistant improvement: improvement:JP,3-122148,A, such as JP,1-318052,A and a moldability) which consists of crystalline polyolefin resin, and ethylene and an annular olefin random copolymer is well-known conventionally as a resin constituent which consists of an annular olefin system copolymer and thermoplastics. However, glass transition (Tg) is 50-230 degrees C, and the random copolymer used with these constituents cannot improve the shock resistance of thermoplastics by blending this random copolymer.
[0005] Hereafter, lessons is taken from this invention and it explains in more detail. First, each component is explained in full detail.
(a) It is the copolymer obtained by copolymerizing an annular olefin and an alpha olefin as an annular olefin system copolymer (a) component, and use the annular olefin system copolymer whose glass

transition temperature (Tg) is 30 degrees C or less. Here, as the above-mentioned alpha olefin, although not necessarily limited, it is the following general formula [X], for example.

[Formula 1]



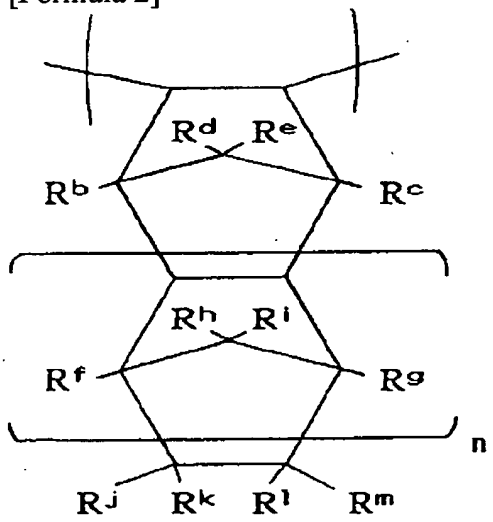
… [X]

(-- Ra shows a hydrogen atom or the hydrocarbon group of carbon numbers 1-20 among a formula [X].) -- what gives the repeat unit expressed is mentioned.

[0006] In the repeat unit shown by the above-mentioned general formula [X], Ra shows the hydrogen atom or the hydrocarbon group of carbon numbers 1-20. Here, specifically, a methyl group, an ethyl group, an isopropyl group, n-propyl group, an isobutyl radical, n-butyl, n-hexyl group, n-octyl radical, n-octadecyl radical, etc. can be mentioned as a hydrocarbon group of carbon numbers 1-20. Moreover, as an example of an alpha olefin of giving the repeat unit shown by the general formula [X], ethylene, a propylene, 1-butene, a 3-methyl-1-butene, 4-methyl-1-pentene, 1-hexene, 1-octene, 1-decene, 1-ray KOSEN, etc. can be mentioned, for example.

[0007] Moreover, as said annular olefin, although not necessarily limited, it is the following general formula [Y], for example.

[Formula 2]



… [Y]

(Rb-Rm shows the substituent containing a hydrogen atom, the hydrocarbon group of carbon numbers 1-20 or a halogen atom, an oxygen atom, or a nitrogen atom among a formula [Y], respectively, and n shows zero or more integers.) Rj, Rk and Rl, or Rm may form a ring mutually. Moreover, even if Rb-Rm is mutually the same respectively, it may differ. What gives the repeat unit expressed is mentioned.

[0008] In the repeat unit expressed with the above-mentioned general formula [Y], Rb-Rm shows the substituent which contains a hydrogen atom, the hydrocarbon group of carbon numbers 1-20 or a halogen atom, an oxygen atom, or a nitrogen atom, respectively. As a hydrocarbon group of carbon numbers 1-20 here specifically For example, the alkyl group of the carbon numbers 1-20, such as a methyl group, an ethyl group, n-propyl group, an isopropyl group, n-butyl, an isobutyl radical, t-butyl, and a hexyl group, The aryl group, alkyl aryl radical, or arylated alkyl radical of the carbon numbers 6-20, such as a phenyl group, a tolyl group, and benzyl, The alkenyl radical of the carbon numbers 2-20, such as an alkylidene radical of the carbon numbers 1-20, such as a methylidyne radical, an ethylidene radical, and a propylidene radical, a vinyl group, and an allyl group, etc. can be mentioned. However, Rb, Rc, Rf, and Rg remove an alkylidene radical. In addition, when Rd, Re, or Rh-Rm is an alkylidene radical, the carbon atom which it has combined does not have other substituents.

[0009] Moreover, the halogenation alkyl group of the carbon numbers 1-20, such as halogen radicals, such as a fluorine, chlorine, a bromine, and iodine, a chloro methyl group, a bromomethyl radical, and a

chloro ethyl group, etc. can specifically as a substituent containing a halogen atom be mentioned. The alkoxy carbonyl group of the carbon numbers 1-20, such as an alkoxy group of the carbon numbers 1-20, such as a methoxy group, an ethoxy radical, a propoxy group, and a phenoxy group, a methoxycarbonyl group, and an ethoxycarbonyl radical, etc. can specifically as a substituent containing an oxygen atom be mentioned. Alkylamino radicals, cyano groups, etc. of carbon numbers 1-20, such as a dimethylamino radical and a diethylamino radical, can specifically as a substituent containing a nitrogen atom be mentioned.

[0010] As an example of an annular olefin of giving the repeat unit shown by the general formula [Y] For example, norbornene, 5-methyl norbornene, 5-ethyl norbornene, 5-propyl norbornene, 5, 6-dimethyl norbornene, 1-methyl norbornene, 7-methyl norbornene, 5 and 5, 6-trimethyl norbornene, 5-phenyl norbornene, 5-benzyl norbornene, 5-ethylidene norbornene, 5-vinyl norbornene, 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-methyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-ethyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2, 3-dimethyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-hexyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-ethylidene - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-fluoro - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 1, 5-dimethyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-cyclohexyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2, 3-dichloro - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 2-isobutyl - 1, 4, 5, 8-dimethano - 1, 2, 3, 4,a [4], 5, 8, and 8a-octahydronaphthalene, 1, a 2-dihydrodicyclopentadiene, 5-chloro norbornene, 5 and 5-dichloro norbornene, 5-fluoro norbornene, 5 and 5, 6-trifluoro-6-trifluoromethyl norbornene, 5-chloro methyl norbornene, 5-methoxy norbornene, 5, 6-dicarboxyl norbornene ANHAIDO rate, 5-dimethylamino norbornene, 5-cyano norbornene, etc. can be mentioned.

[0011] Fundamentally, although the annular olefin system copolymer used by this invention comes to copolymerize an alpha olefin and an annular olefin which were mentioned above, it is the range which does not spoil the purpose of this invention, and other copolymerizable partial saturation monomer components other than these indispensable 2 components may be used for it if needed. As a partial saturation monomer by which copolymerization may be carried out to such arbitration, specifically ** What is not previously used among the above mentioned alpha olefin components, ** What is not previously used among the above mentioned annular olefin components, ** Monocycle olefins, such as chain-like dienes, such as annular diene [, such as a dicyclopentadiene and norbornadiene,], ** butadiene, isoprene, 1, and 5-hexadiene, ** cyclopentene, and a cyclo heptene, are mentioned.

[0012] An annular olefin system copolymer requires that glass transition temperature (Tg) should be 30 degrees C or less. If such a copolymer is used, the impact-proof of thermoplastics and a heat-sealing property are improvable. -30-30 degrees C (Tg) especially of more desirable glass transition temperature are -30-20 degrees C. In this case, by changing the class of monomer, and a presentation, the annular olefin system copolymer used by this invention can control glass transition temperature (Tg) to arbitration, and can change glass transition temperature (Tg) into arbitration according to the application of the constituent made into the purpose, other thermoplastics, the temperature used.

[0013] Furthermore, as for an annular olefin system copolymer, it is desirable that 90 degrees C or less of melting points by DSC are 20-80 degrees C especially also in 10-85 degrees C. In this case, it is suitable for an annular olefin system copolymer that the broadcloth fusion peak by DSC is less than 90 degrees C. The improvement effectiveness of the constituent obtained, such as shock resistance, may become the presentation distribution of the copolymer of an annular olefin and an alpha olefin of a copolymer which has a sharp fusion peak by DSC in 90 degrees C or more is uneven, and inadequate. DSC measurement -- it is, and if the melting point (fusion peak) of the annular olefin system copolymer used by this invention is not seen by Sharp but is one of the things of low crystallinity especially, on the Measuring condition level of usual polyethylene, a peak hardly comes out of it. Furthermore, as for an annular olefin system copolymer, in the crystallization peak by DSC (temperature fall measurement), it

is desirable that it is what has a comparatively small subpeak (more than a piece) in the elevated-temperature side of the Maine peak. According to the description of these thermal properties, the improvement effectiveness of a constituent, such as shock resistance, is acquired in large operating temperature limits.

[0014] In the annular olefin system copolymer used by this invention, the content [x] of the repeat unit originating in an alpha olefin and the content [y] of the repeat unit originating in an annular olefin comparatively ([x] : [y]) It changes with the classes and combination of an alpha olefin and an annular olefin. although it cannot necessarily perform specifying generally -- usually -- 80-99.9 mol %:20 - 0.1-mol % -- desirable -- 82-99.5 mol %:18 - 0.5-mol % -- it is 85-98 mol %:15 - two-mol % especially preferably. When the content [y] of an annular olefin repeat unit exceeds 20-mol %, the glass transition point of a copolymer and a modulus of elasticity in tension may become high, and may become inadequate [the shock resistance of a constituent and mold goods, and toughness], and when it is less than [0.1 mol %], the degree of crystallinity of a copolymer becomes high too much, and may become too inadequate [the shock resistance of a constituent and mold goods etc.].

[0015] Moreover, as an annular olefin system copolymer, the repeat unit originating in an alpha olefin and the repeat unit originating in an annular olefin are the copolymers of the parenchyma top line arranged in the shape of a straight chain, and it is desirable that it is what does not have the gel structure of cross linkage. When a copolymer dissolves completely into a 135-degree C decalin, it can check not having the gel structure of cross linkage.

[0016] As for an annular olefin system copolymer, it is desirable that the limiting viscosity [eta] measured in the 135-degree C decalin is 0.01 - 20 dl/g. When limiting viscosity [eta] is less than 0.01 dl/g, the reinforcement of mold goods may fall, and when 20 dl/g is exceeded, the moldability of a constituent may worsen. More desirable limiting viscosity [eta] is 0.05 - 10 dl/g.

[0017] Moreover, although especially the molecular weight of an annular olefin system copolymer is not restricted, for the weight average molecular weight Mw measured with gel permeation chromatography (GPC), 1,000-2,000,000 especially 5,000-1,000,000, and number average molecular weight Mn are 500-1,000,000, especially 2,000-800,000, and 1.3-4, and that it is especially 1.4-3 have molecular weight distribution (Mw/Mn) desirable [molecular weight]. When molecular weight distribution (Mw/Mn) become larger than 4, the content of a low-molecular-weight object may increase, and it may become the cause which carries out bleed out from mold goods.

[0018] Moreover, as for an annular olefin system copolymer, it is desirable that the degree of crystallinity measured with the X-ray diffraction method is 0 - 40%. When degree of crystallinity exceeds 40%, the shock resistance of mold goods may become inadequate. Especially more desirable degree of crystallinity is 0 - 25% 0 to 30%.

[0019] Moreover, as for the annular olefin system copolymer used by this invention, it is desirable that moduli of elasticity in tension are two or less 3,000 kg/cm. When a modulus of elasticity in tension exceeds 3,000 kg/cm2, the toughness of mold goods may become inadequate. More desirable moduli of elasticity in tension are 50 - 2,000 kg/cm2, especially 100 - 1,500 kg/cm2.

[0020] although it has the physical properties of the range mentioned above as an annular olefin system copolymer used by this invention -- Mika and others -- you may be a copolymer and may be the mixture of the copolymer which has different Tg whose glass transition temperature (Tg) is 30 degrees C or less in this case. Moreover, a part of copolymer which has the physical properties outside the above-mentioned range may be contained. In the case of the latter, the whole physical-properties value should just be included in the above-mentioned range.

[0021] Although there is no limitation in the manufacture approach of the annular olefin system copolymer used by this invention, it can manufacture efficiently by performing copolymerization of an alpha olefin and an annular olefin using the catalyst which uses as a principal component the catalyst which uses the following compound (A) and (B) as a principal component or the following compound (A), (B), and (C).

(A) The compound (C) organoaluminium compound which reacts with the transition-metals (compound B) transition-metals compound, and forms the complex of ionicity [0022] In this case, the transition-

metals compound which contains the transition metals belonging to the IVB group of the periodic table, VB group, a VIB group, a VIIB group, and a VIII group as the above-mentioned transition-metals compound (A) can be used. As the above-mentioned transition metals, titanium, a zirconium, a hafnium, chromium, manganese, nickel, palladium, platinum, etc. are desirable, and, specifically, a zirconium, a hafnium, titanium, nickel, and palladium are desirable especially.

[0023] As such a transition-metals compound (A), although various things are mentioned The compound which contains the transition metals of an IVB group and a VIII group especially, the transition metals chosen from the IVB group of the periodic table especially, Namely, the compound containing titanium (Ti), a zirconium (Zr), or a hafnium (Hf) can be used suitably. The compounds especially shown by the following general formula (I), (II), the cyclopentadienyl compounds that are shown, these derivatives, or the following general formula (IV), or these derivatives are suitable.

CpM1R1aR2bR3c -- (I)

Cp2M1R1aR2b -- (II)

(Cp-Ae-Cp) M1R1aR2b -- (III)

M1R1aR2bR3cR4d -- (IV)

[0024] M1 shows Ti, Zr, or Hf atom among a [(I) - (IV) type, and Cp shows annular unsaturated hydrocarbon radicals, such as a cyclopentadienyl group, a substituent cyclopentadienyl group, an indenyl group, a permutation indenyl group, a tetrahydro indenyl group, a permutation tetrahydro indenyl group, a fluorenyl group, or a permutation fluorenyl group, or a chain-like unsaturated hydrocarbon radical. R1, R2, and R3 and R4 show ligands, such as a ligand of sigma bond nature, a chelated ligand, and a Lewis base, respectively. As a ligand of sigma bond nature Concretely A hydrogen atom, an oxygen atom, a halogen atom, the alkyl group of carbon numbers 1-20, the alkoxy group of carbon numbers 1-20, the aryl group of carbon numbers 6-20, an alkyl aryl radical or an arylated alkyl radical, the acyloxy radical of carbon numbers 1-20, an allyl group, a permutation allyl group, The substituent containing a silicon atom etc. can be illustrated and an acetylacetonato radical, a permutation acetylacetonato radical, etc. can be illustrated as a chelated ligand. A shows bridge formation by covalent bond. a, b, c, and d show the integer of 0-4, and e shows the integer of 0-6, respectively. R1, R2, and R3 and R4 -- the -- two or more may join together mutually and it may form the ring. When Above Cp has a substituent, the substituent concerned has the desirable alkyl group of carbon numbers 1-20. (II) In a formula and (III) a formula, two Cp(s) may be the same and may differ mutually.]

[0025] As a substituent cyclopentadienyl group in an above (I) - (III) type, a methylcyclopentadienyl radical, an ethylcyclopentadienyl radical, an isopropyl cyclopentadienyl group, 1, 2-dimethylcyclopentadienyl radical, a tetramethylcyclopentadienyl radical, 1, 3-dimethylcyclopentadienyl radical, 1 and 2, 3-trimethylcyclopentadienyl radical, 1 and 2, 4-trimethylcyclopentadienyl radical, a pentamethylcyclopentadienyl group, a trimethylsilylcyclopentadienyl radical, etc. are mentioned, for example. moreover, as an example of R1-R4 in an above (I) - (IV) type For example As a halogen atom, as an alkyl group of a fluorine atom, a chlorine atom, a bromine atom, and the iodine atom; carbon numbers 1-20 A methyl group, an ethyl group, n-propyl group, an iso-propyl group, n-butyl, an octyl radical, a 2-ethylhexyl radical; as an alkoxy group of carbon numbers 1-20 A methoxy group, an ethoxy radical, a propoxy group, a butoxy radical, a phenoxy group; as the aryl group, alkyl aryl radical, or arylated alkyl radical of carbon numbers 6-20 A phenyl group, a tolyl group, a xylyl group, benzyl; as an acyloxy radical of carbon numbers 1-20 As a substituent containing a silicon atom, as a trimethylsilyl radical and a methyl group (trimethylsilyl):Lewis base Heptadecyl carbonyloxy group; Ether, such as wood ether, diethylether, and a tetrahydrofuran, Ester, such as thioether, such as tetrahydrothiophene, and ethyl benzoate Amines, such as a nitril [, such as an acetonitrile and a benzonitrile,], trimethylamine, triethylamine, tributylamine, N.N-dimethylaniline, pyridine, 2, and 2'-bipyridine and a phenanthroline, Phosphines, such as a triethyl phosphine and triphenyl phosphine; as chain-like unsaturated hydrocarbon Ethylene, a butadiene, 1-pentene, an isoprene, pentadiene, 1-hexenes, and these derivatives; as annular unsaturated hydrocarbon Benzene, toluene, a xylene, cycloheptatriene, cyclo-octadiene, cyclooctatriene, cyclo-octatriene, these derivatives, etc. are mentioned. Moreover, as bridge

formation by the covalent bond of A in the above-mentioned (III) formula, methylene bridge formation, dimethyl methylene bridge formation, ethylene bridge formation, 1, and 1'-cyclo hexylene bridge formation, dimethyl silylene bridge formation, a dimethylgermirene crosslinking, dimethyl SUTANIREN bridge formation, etc. are mentioned, for example.
[0026] As such a compound, the compound which permuted the following and the zirconium of these compounds with titanium or a hafnium is mentioned.
(I) The compound (pentamethylcyclopentadienyl) trimethyl zirconium of a formula, A triphenyl zirconium, (Pentamethylcyclopentadienyl) (pentamethylcyclopentadienyl)tribenzylzirconium, A TORIKURORO zirconium, a trimethoxy (pentamethylcyclopentadienyl) zirconium, (Pentamethylcyclopentadienyl) A trimethyl zirconium, a triphenyl (cyclopentadienyl) zirconium, (Cyclopentadienyl) (cyclopentadienyl)tribenzylzirconium, a TORIKURORO (cyclopentadienyl) zirconium, A trimethoxy zirconium, a dimethyl (cyclopentadienyl) (methoxy) zirconium, (Cyclopentadienyl) A trimethyl zirconium, a triphenyl (methylcyclopentadienyl) zirconium, (Methylcyclopentadienyl) (methylcyclopentadienyl)tribenzylzirconium, a TORIKURORO (methylcyclopentadienyl) zirconium, A dimethyl (methoxy) zirconium, (Methylcyclopentadienyl) A TORIKURORO zirconium, a TORIKURORO (trimethylcyclopentadienyl) zirconium, (Dimethylcyclopentadienyl) (Trimethylsilylcyclopentadienyl) A trimethyl zirconium, a TORIKURORO (tetramethylcyclopentadienyl) zirconium, [0027] (II) The compound bis(cyclopentadienyl) dimethyl zirconium of a formula, A bis(cyclopentadienyl) diphenyl zirconium, a bis(cyclopentadienyl) diethyl zirconium, A bis(cyclopentadienyl) dibenzyl zirconium, a bis(cyclopentadienyl) dimethoxy zirconium, A bis(cyclopentadienyl) dichloro zirconium, a bis(cyclopentadienyl) dihydrido zirconium, A bis (cyclopentadienyl) monochloro mono-hydride zirconium, A bis(methylcyclopentadienyl) dimethyl zirconium, a bis(methylcyclopentadienyl) dichloro zirconium, A bis(methylcyclopentadienyl) dibenzyl zirconium, a bis(pentamethylcyclopentadienyl) dimethyl zirconium, A bis (pentamethylcyclopentadienyl) dichloro zirconium, A bis(pentamethylcyclopentadienyl) dibenzyl zirconium, A bis(pentamethylcyclopentadienyl) chloro methyl zirconium, a bis (pentamethylcyclopentadienyl) hydride methyl zirconium, a dichloro (pentamethylcyclopentadienyl (cyclopentadienyl)) zirconium, [0028] The compound ethylene bis(indenyl) dimethyl zirconium of a formula, (III) An ethylene bis(indenyl) dichloro zirconium, an ethylene bis(tetrahydro indenyl) dimethyl zirconium, An ethylene bis(tetrahydro indenyl) dichloro zirconium, a dimethyl silylene bis (cyclopentadienyl) dimethyl zirconium, A dimethyl silylene bis(cyclopentadienyl) dichloro zirconium, An isopropylidene (cyclopentadienyl) (9-fluorenyl) dimethyl zirconium, An isopropylidene (cyclopentadienyl) (9-fluorenyl) dichloro zirconium, A [phenyl (methyl) methylene] (9-fluorenyl) (cyclopentadienyl) dimethyl zirconium, A diphenylmethylene (cyclopentadienyl) (9-fluorenyl) dimethyl zirconium, An ethylene (9-fluorenyl) (cyclopentadienyl) dimethyl zirconium, A cyclohexylidene (9-fluorenyl) (cyclopentadienyl) dimethyl zirconium, A cyclo pen dust DIN (9-fluorenyl) (cyclopentadienyl) dimethyl zirconium, A cyclo butylidene (9-fluorenyl) (cyclopentadienyl) dimethyl zirconium, A dimethyl silylene (9-fluorenyl) (cyclopentadienyl) dimethyl zirconium, A dimethyl silylene bis(2, 3, 5-trimethylcyclopentadienyl) dichloro zirconium, A dimethyl silylene bis(2, 3, 5-trimethylcyclopentadienyl) dimethyl zirconium, a dimethyl silylene SUBISU (indenyl) dichloro zirconium [0029] A zirconium compound with one sort of alkyl groups, such as a compound which the compound of the aforementioned (IV) formula was mentioned, for example, transposed the following compounds or these zirconiums to a hafnium and titanium as an example of compounds other than the cyclopentadienyl compound shown by the above-mentioned general formula (I), (II), and (III), an alkoxy group, and a halogen atom, or two sorts or more, a hafnium compound, and a titanium compound are mentioned.
A tetramethyl zirconium, a tetra-benzyl zirconium, a tetramethoxy zirconium, a tetra-ethoxy zirconium, tetra-butoxyzirconium, a tetra-chloro zirconium, a tetrabromo zirconium, a butoxy TORIKURORO zirconium, a dibutoxy dichloro zirconium, a bis(2, 5-G t-butyl phenoxy) dimethyl zirconium, a bis(2, 5-G t-butyl phenoxy) dichloro zirconium, a zirconium screw (acetylacetonato), [0030] moreover, as a transition-metals compound containing a VB-VIII group's transition metals There is especially no limit.

As an example of a chromium compound For example, tetramethyl chromium, Tetrapod (t-butoxy) chromium, bis(cyclopentadienyl) chromium, Hydride tricarbonyl (cyclopentadienyl) chromium, hexa carbonyl (cyclopentadienyl) chromium, Bis(benzene) chromium, tricarbonyl tris (phosphonic acid triphenyl) chromium, tris (allyl compound) chromium, triphenyl tris (tetrahydrofuran) chromium, chromium tris (acetylacetonato), etc. are mentioned.

[0031] As an example of a manganese compound, tricarbonyl (cyclopentadienyl) manganese, PENTA carbonylmethyl manganese, bis(cyclopentadienyl) manganese, a manganese screw (acetylacetonato), etc. are mentioned, for example.

[0032] As an example of a nickel compound, for example Dicarbonyl bis(triphenyl phosphine) nickel, Dibromobis (triphenyl phosphine) nickel, a dinitrogen screw (bis(tri-cyclohexyl phosphine) nickel), Chloro hydride bis(tri-cyclohexyl phosphine) nickel, chloro (phenyl) bis(triphenyl phosphine) nickel, Dimethyl bis(trimethyl phosphine) nickel, diethyl (2 and 2'-bipyridyl) nickel, Bis(allyl compound) nickel, bis(cyclopentadienyl) nickel, Bis(methylcyclopentadienyl) nickel, bis (pentamethylcyclopentadienyl) nickel, Allyl compound (cyclopentadienyl) nickel, nickel (cyclo-octadiene (cyclopentadienyl)) tetrafluoroborate, Bis(cyclo-octadiene) nickel, nickel bisacetylacetonate, Allyl compound nickel chloride, tetrakis (triphenylphosphine) nickel, A nickel chloride, nickel (C6H5) {OC(C6H5) CH=P (C6H5)2}, {P(C6H5) 3}, nickel (C6H5) {OC(C6H5) C(SO3Na) =P(C6H5) 2}, {P (C6H5) 3}, etc. are mentioned.

[0033] As an example of a palladium compound, for example Dichlorobis (benzonitrile) palladium, Carbonyl tris (triphenyl phosphine) palladium, dichlorobis (triethyl phosphine) palladium, Bis(iso cyanidation t-butyl) palladium, a palladium screw (acetylacetonato), Dichloro (tetra-phenyl cyclobutadiene) palladium, dichloro (1, 5-cyclo-octadiene) palladium, Allyl compound (cyclopentadienyl) palladium, bis(allyl compound) palladium, Allyl compound (1, 5-cyclo-octadiene) palladium tetrafluoroborate, palladium (1 (acetylacetonato), 5-cyclo-octadiene) tetrafluoroborate, tetrakis (acetonitrile) palladium 2 tetrafluoroborate, etc. are mentioned.

[0034] Next, although anything can be used if it is the compound which reacts with a transition-metals compound (A) and forms the complex of ionicity as a compound (B), the compound with which a cation and two or more radicals consist of an anion combined with the element, especially the coordination complex compound with which a cation and two or more radicals consist of an anion combined with the element can be used suitably. As a compound with which such a cation and two or more radicals consist of an anion combined with the element, the compound shown by the following formula (V) or (VI) can be used suitably.

([L1-R7] k+) p([M3Z1Z2 --Zn] (n-m)-) q -- (V)

([L2] k+) p([M4Z1Z2 --Zn] (n-m)-) q -- (VI)

(However, L2 is M5, R8R9M6, R103C, or R11M6)

[0035] The inside of [(V) and the (VI) type, the element with which a Lewis base, and M3 and M4 are chosen for L1 from VB group, the VIB group, the VIIB group, the VIII group, IB group, the IIB group, the IIIA group, IVA group, and VA group of the periodic table, respectively, The element preferably chosen from an IIIA group, an IVA group, and VA group, the element with which M5 and M6 are chosen from the IIIB group, the IVB group, VB group, the VIB group, the VIIB group, the VIII group, IA group, IB group, the IIA group, IIB group, and VIIA group of the periodic table, respectively, Z1-Zn A ********** hydrogen atom, a dialkylamino radical, the alkoxy group of carbon numbers 1-20, the aryloxy group of carbon numbers 6-20, the alkyl group of carbon numbers 1-20, the aryl group of carbon numbers 6-20, an alkyl aryl radical, an arylated alkyl radical, the halogenation hydrocarbon group of carbon numbers 1-20, the acyloxy radical, the organic metalloid radical, or halogen atom of carbon numbers 1-20 -- being shown -- Z1-Zn -- the -- two or more may join together mutually and it may form the ring. R7 shows a hydrogen atom, the alkyl group of carbon numbers 1-20, the aryl group of carbon numbers 6-20, an alkyl aryl radical, or an arylated alkyl radical, and, as for a cyclopentadienyl group, a substituent cyclopentadienyl group, an indenyl group or a fluorenyl group, and R10, R8 and R9 show the alkyl group, the aryl group, the alkyl aryl radical, or arylated alkyl **** of carbon numbers 1-20, respectively. R11 shows large annular ligands, such as a tetra-phenyl porphyrin and a

phthalocyanine. For m, the integer of 1-7 and n are [the integer of 1-7 and p of the integer of 2-8 and k] one or more integers and q=(pxk)/(n-m) in the ionic valence of [L1-R7] and [L2] at the valence of M3 and M4.]

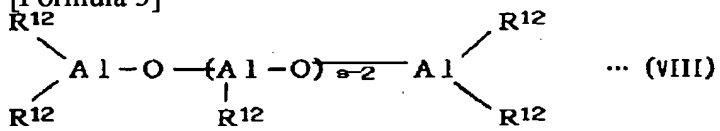
[0036] As an example of the above-mentioned Lewis base Ammonia, monomethylamine, an aniline, dimethylamine, diethylamine, N-methylaniline, a diphenylamine, a trimethylamine, triethylamine, tree n butylamine, N.N-dimethylaniline, a methyl diphenylamine, a pyridine, p-promo-N, N-dimethylaniline, p-nitro -Amines, such as N and N-dimethylaniline, Phosphoretted hydrogen, such as triethyl phosphoretted hydrogen, triphenylphosphine, and diphenyl phosphoretted hydrogen Ester, such as thioether, such as ether, such as wood ether, diethylether, a tetrahydrofuran, and dioxane, a diethyl thioether, and tetrahydrothiophene, and ethyl BENZOTO, is mentioned. as the example of M3 and M4 -- B, aluminum, Si, P, As, Sb, etc. -- as B or an example of P and M5, Mn, Fe, Co, nickel, Zn, etc. are preferably mentioned as Li, Na, Ag, Cu, Br, I, and I3 grade and an example of M6.

[0037] Z1 - Zn As an example, for example As a dialkylamino radical, as a dimethylamino radical and an ARUKOSHIKI radical of the diethylamino radical; carbon numbers 1-20 A methoxy group, an ethoxy radical, an n-butoxy radical; as an aryloxy group of carbon numbers 6-20 A phenoxy group, 2, 6-dimethyl phenoxy group, a naphthyloxy radical; as an alkyl group of carbon numbers 1-20 A methyl group, an ethyl group, n-propyl group, an iso-propyl group, n-butyl, n-octyl radical, a 2-ethylhexyl radical; as the aryl group, alkyl aryl radical, or arylated alkyl radical of carbon numbers 6-20 A phenyl group, p-tolyl group, benzyl, a 4-tertiary-buthylphenyl radical, 2, 6-dimethylphenyl radical, 3, 5-dimethylphenyl radical, 2, 4-dimethylphenyl radical, 2, 3-dimethylphenyl radical; as a halogenation hydrocarbon group of carbon numbers 1-20 p-fluoro phenyl group, 3, a 5-difluoro phenyl group, a pentachlorophenyl group, 3 and 4, 5-trifluoro phenyl group, a pentafluorophenyl radical, 3, 5-JI (trifluoromethyl) phenyl group; as a halogen atom F, Cl, Br, I; a 5 methyl antimony radical, a trimethylsilyl radical, a trimethyl gel mill radical, a diphenyl arsine radical, a dicyclohexyl antimony radical, and a diphenyl boron radical are mentioned as an organic metalloid radical. What was mentioned previously, and the same thing are mentioned as an example of R7 and R10. As an example of the substituent cyclopentadienyl group of R8 and R9, what was permuted by alkyl groups, such as a methylcyclopentadienyl radical, a butylcyclopentadienyl radical, and a pentamethylcyclopentadienyl group, is mentioned. Here, the number of alkyl groups with which alkyl groups are 1-6, and the carbon number was permuted can usually be chosen for the integer of 1-5. In (V) and (VI)-type compound, that M3 and whose M4 are boron is desirable.

[0038] Specifically, the following can be especially used suitably in (V) and (VI)-type compound. (V) Compound tetra-phenyl triethyl borate ammonium of a formula, tetra-phenyl boric acid Tori (n-butyl) ammonium, Tetra-phenyl boric acid trimethylammonium, tetra-phenyl boric acid tetraethylammonium, Tetra-phenyl methyl borate Tori (n-butyl) ammonium, tetra-phenyl boric acid benzoRUTORI (n-butyl) ammonium, Tetra-phenyl boric acid dimethyl diphenyl ammonium, tetra-phenyl methyl borate triphenyl ammonium, Tetra-phenyl trimethyl borate anilinium, tetra-phenyl methyl borate pyridinium, Tetra-phenyl boric acid benzyl pyridinium, tetra-phenyl methyl borate (2-cyano pyridinium), tetra-phenyl trimethyl borate sulfonium, tetra-phenyl boric acid benzyl dimethyl sulfonium,

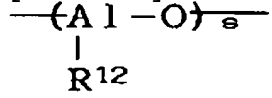
[0039] Tetrakis (pentafluorophenyl) triethyl borate ammonium, Tetrakis (pentafluorophenyl) boric acid Tori (n-butyl) ammonium, Tetrakis (pentafluorophenyl) boric acid triphenyl ammonium, Tetrakis (pentafluorophenyl) boric acid tetrabutylammonium, Tetrakis (pentafluorophenyl) boric acid (tetraethylammonium), Tetrakis (pentafluorophenyl) boric acid (MECHIRUTORI (n-butyl) ammonium), Tetrakis (pentafluorophenyl) boric acid (benzoRUTORI (n-butyl) ammonium), Tetrakis (pentafluorophenyl) methyl borate diphenyl ammonium, Tetrakis (pentafluorophenyl) methyl borate triphenyl ammonium, Tetrakis (pentafluorophenyl) boric acid dimethyl diphenyl ammonium, Tetrakis (pentafluorophenyl) boric acid anilinium, tetrakis (pentafluorophenyl) methyl borate anilinium, Tetrakis (pentafluorophenyl) boric acid dimethyl anilinium, Tetrakis (pentafluorophenyl) trimethyl borate anilinium, tetrakis (pentafluorophenyl) boric acid dimethyl (m-nitro anilinium), tetrakis (pentafluorophenyl) boric acid dimethyl (p-BUROMO anilinium), [0040] Tetrakis (pentafluorophenyl) boric acid pyridinium, tetrakis (pentafluorophenyl) boric acid (p-cyano pyridinium), Tetrakis

(pentafluorophenyl) boric acid (N-methyl pyridinium), Tetrakis (pentafluorophenyl) boric acid (N-benzyl pyridinium), Tetrakis (pentafluorophenyl) boric acid (O-cyano-N-methyl pyridinium), Tetrakis (pentafluorophenyl) boric acid (p-cyano-N-methyl pyridinium), Tetrakis (pentafluorophenyl) boric acid (p-cyano-N-benzyl pyridinium), Tetrakis (pentafluorophenyl) trimethyl borate sulfonium, Tetrakis (pentafluorophenyl) boric acid benzyl dimethyl sulfonium, Tetrakis (pentafluorophenyl) boric acid tetra-phenyl phosphonium, Tetrakis (pentafluorophenyl) boric acid triphenyl phosphonium, tetrakis (3, 5-ditrifluoromethyl phenyl) boric acid dimethyl anilinium, hexafluoro arsenic acid triethyl ammonium,
[0041] (VI) Compound tetra-phenyl boric acid ferro SENIUMU of a formula, tetra-phenyl boric acid silver, Tetra-phenyl boric acid trityl, tetra-phenyl boric acid (tetra-phenyl porphyrin manganese), Tetrakis (pentafluorophenyl) boric acid ferro SENIUMU, tetrakis (pentafluorophenyl) boric acid (1 and 1'-dimethyl ferro SENIUMU), Tetrakis (pentafluorophenyl) boric acid decamethyl ferro SENIUMU, Tetrakis (pentafluorophenyl) boric acid acetyl ferro SENIUMU, Tetrakis (pentafluorophenyl) boric acid formyl ferro SENIUMU, Tetrakis (pentafluorophenyl) boric acid cyano ferro SENIUMU, Tetrakis (pentafluorophenyl) boric acid silver, tetrakis (pentafluorophenyl) boric acid trityl, Tetrakis (pentafluorophenyl) lithium borate, tetrakis (pentafluorophenyl) sodium borate, Tetrakis (pentafluorophenyl) boric acid (tetra-phenyl porphyrin manganese), Tetrakis (pentafluorophenyl) boric acid (tetra-phenyl porphyrin iron chloride), Tetrakis (pentafluorophenyl) boric acid (tetra-phenyl porphyrin zinc), silver tetrafluoborate, silver hexafluoroarsenate, hexafluoro antimonic acid silver,
[0042] Moreover, compounds other than (V) and the (VI) type, for example, Tori (pentafluorophenyl) boron, Tori (3, 5-JI (trifluoromethyl) phenyl) boron, triphenyl boron, etc. are usable.
[0043] (C) As an organoaluminium compound which is a component, the following general formula (VII) or (VIII) the thing shown by (IX) is mentioned.
R12rAlQ3-r -- (VII)
R12 -- carbon numbers 1-20 -- hydrocarbon groups, such as an alkyl group of 1-12, an alkenyl radical, an aryl group, and an arylated alkyl radical, and Q show the alkoxy group or halogen atom of a hydrogen atom and carbon numbers 1-20 preferably. r is the thing of the range of $1<=r<=3$.
Specifically as a compound of a formula (VII), trimethylaluminum, triethylaluminum, triisopropyl aluminum, triisobutylaluminum, dimethyl aluminum chloride, a diethylaluminium chloride, methyl aluminum dichloride, ethyl aluminum dichloride, a dimethyl aluminum fluoride, a diisobutyl aluminum hydride, a diethyl aluminum hydride, ethylaluminium sesquichloride, etc. are mentioned.
[0044]
[Formula 3]



… (VIII)

(R12 shows the same thing as a formula (VII).) s -- polymerization degree -- being shown -- usually -- 3-50 -- it is 7-40 preferably. Chain-like aluminoxane shown.
[0045]
[Formula 4]



… (IX)

(R12 shows the same thing as a formula (VII).) moreover, s -- polymerization degree -- being shown -- the desirable repeat number of unit -- 3-50 -- it is 7-40 preferably. Annular alkyl aluminoxane which has the repeat unit shown. A desirable compound of a (VII)- (IX) type is a with a carbon numbers of three or more alkyl group, the alkyl group content aluminium compound which has at least one or more branching alkyl groups especially, or aluminoxane. Especially a desirable thing is triisobutylaluminum or with a polymerization degree of seven or more aluminoxane. High activity can be acquired when this triisobutylaluminum, with a polymerization degree of seven or more aluminoxane, or such mixture are

used. Moreover, insoluble denaturation aluminoxane can be used for the usual solvent which denaturalized the aluminoxane shown by the (VIII)- (IX) formula with the compound with active hydrogen, such as water.

[0046] What is necessary is for there to be especially no limitation in the means, and just to make it react to it according to a well-known approach, although the method of contacting alkylaluminum and condensing agents, such as water, is mentioned as a manufacturing method of said aluminoxane. For example, there is an approach to which dissolve ** organoaluminium compound in the organic solvent, add the organoaluminium compound at the beginning at the time of the approach of contacting this in water, and ** polymerization, make trialkylaluminium react to the approach and ** tetra-ARUKIRUJI aluminoxane to which the water of crystallization contained in the approach of adding water behind, ** metal salt, etc. and the water of adsorption to an inorganic substance or the organic substance are made to react with an organoaluminium compound, and water is made to react further.

[0047] The catalyst used for manufacture of an annular olefin system copolymer uses the above-mentioned (A) component, the (B) component or the above-mentioned (A) component and the (B) component, and the (C) component as a principal component. In this case, although the service condition of the (A) component and the (B) component is not limited, it is desirable 1:0.01-1:100 and to set the ratio (mole ratio) of a (A) component:(B) component to 1:1-1:5 especially also in 1:0.5-1:10. Moreover, as for service temperature, it is desirable to consider as the range of -100-250 degrees C, and a pressure and time amount can be set as arbitration.

[0048] Moreover, the 5-1,000 mols of the amount of the (C) component used are 0-2,000 mols usually 10-500 mols especially preferably preferably to one mol of (A) components. (C) If a component is used, improvement in polymerization activity can be aimed at, but an organoaluminium compound remains [so much] in a polymer and is not desirable if many [not much]. In addition, there is no limit in the use mode of a catalyst, for example, (A) and the (B) component are contacted beforehand, or a contact product may be used further, separating and washing it, and you may use it, making it contact within a polymerization system. Moreover, the (C) component may be beforehand contacted to the contact product of the (A) component, the (B) component or the (A) component, and the (B) component, and may be used. Contact may be contacted beforehand and may be contacted within a polymerization system. Furthermore, a catalyst component can be beforehand added to a monomer and a polymerization solvent, or can also be added in a polymerization system.

[0049] As a polymerization method, which approaches, such as a bulk polymerization, solution polymerization, a suspension polymerization, and a vapor phase polymerization, may be used. Moreover, a batch method or a continuous magnetization method may be used. It is desirable to make -100-250 degrees C especially of polymerization temperature into -50-200 degrees C about polymerization conditions. Moreover, as for the operating rate of a catalyst to a reaction raw material, it is desirable 1-109, and that a raw material monomer / the above-mentioned (A) component (mole ratio), or a raw material monomer / the above-mentioned (B) component (mole ratio) is set especially to 100-107. furthermore, polymerization time amount -- usually -- 1 minute - 10 hours and reaction pressure -- ordinary pressure -100 kg/cm2G -- it is ordinary pressure -50 kg/cm2G preferably. As the accommodation approach of the molecular weight of a polymer, it can be based on selection of the amount of each catalyst component used, and polymerization temperature, and a pan at the polymerization reaction under hydrogen existence.

[0050] When using a polymerization solvent, halogenated hydrocarbon, such as aliphatic hydrocarbon, such as alicyclic hydrocarbon, such as aromatic hydrocarbon, such as benzene, toluene, a xylene, and ethylbenzene, a cyclopentane, a cyclohexane, and a methylcyclohexane, a pentane, a hexane, a heptane, and an octane, chloroform, and dichloromethane, etc. can be used. These solvents may use one sort independently and may combine two or more sorts of things. Moreover, monomers, such as an alpha olefin, may be used as a solvent.

[0051] (b) Although there is especially no limitation in the class of thermoplastics used as a thermoplastics (b) component, it is the alpha olefin system homopolymer or copolymer which uses as one component the alpha olefin expressed with following general formula CH2=CHR13 (a hydrogen

atom or the alkyl group of carbon numbers 1-20 is shown by the inside R13 of a formula.) at least, for example, and things other than said annular olefin system copolymer can be used.
[0052] As such thermoplastics, specifically Polyethylene, such as high density polyethylene, low density polyethylene, and straight chain low density polyethylene, Ethylene and 1-butene copolymer, ethylene and a 4-methyl-1-pentene copolymer, Ethylene and 1-hexene copolymer, ethylene and an annular olefine copolymer (Tg exceeds 30 degrees C), An ethylene-vinylacetate copolymer, an ethylene acrylic-acid copolymer, and its metal salt, Polypropylene, ethylene propylene rubber, a propylene and 1-butene copolymer, Poly1 butene, 1-butene ethylene copolymer, 1-butene propylene copolymer, 1-butene and a 4-methyl-1-pentene copolymer, poly4 methyl 1 pentene, a Pori 3-methyl-1-butene, etc. are raised.
[0053] Moreover, if it considers as thermoplastics, polystyrene, ABS plastics, an AS resin, polyvinyl alcohol, a polymethyl methacrylate, a polyvinyl chloride, a polyvinylidene chloride, fluororesins (polytetrafluoroethylene etc.), a polycarbonate, polyarylate, polyethylene terephthalate, polybutylene terephthalate, polyphenylene sulfide, polyether sulphone, a polyamide, polyimide, polyphenylene oxide, polyacetal, etc. can be used. Especially desirable things are HDPE, LDPE, L-LDPE, polypropylene, ethylene propylene rubber, an annular olefin ring-opening-polymerization object, polystyrene, a polyvinyl chloride, etc. as thermoplastics. In addition, thermoplastics can use two or more sorts together as occasion demands.
[0054] the thermoplastics constituent of this invention -- setting -- the loadings of the (a) component -- 2 - 98% of the weight of the whole -- desirable -- 5 - 95 % of the weight -- especially -- desirable -- 10 - 90 % of the weight -- it is -- the loadings of the (b) component -- whole 98- it is 90 - 10 % of the weight especially preferably 95 to 5% of the weight preferably 2% of the weight. (a) If the loadings of a component exceed 98 % of the weight, the property of thermoplastics will become inadequate, and if the loadings of the (b) component exceed 98 % of the weight, the improvement effectiveness, such as shock resistance, will become inadequate.
[0055] An elastomer can be blended with the resin constituent of arbitration component this invention if needed, and the effectiveness that the shock resistance in low temperature improves especially by this can be acquired to it. Here, although there is no limit in the class of elastomer, thermoplastic elastomer and other rubber can be used suitably. Specifically as thermoplastic elastomer, thermoplastic polyurethane, a styrene butadiene block copolymer, a styrene isoprene block copolymer, etc. are mentioned. Specifically as other rubber, natural rubber, styrene-butadiene rubber, butadiene rubber, polyisoprene rubber, chloroprene rubber, isobutylene isoprene rubber, ethylene-propylene rubber (EPM), EPDM, chlorination polystyrene rubber, etc. are mentioned. Especially desirable things are ethylene-propylene rubber (EPM), EPDM, styrene-butadiene rubber, etc. as an elastomer. When blending an elastomer, as for especially the loadings, it is desirable to carry out to below the 100 weight sections more than 5 weight sections below the 200 weight sections more than 2 weight sections to the thermoplastics 100 weight section. If there are few loadings of an elastomer than 2 weight sections, the improvement effectiveness of low-temperature impact resistance will become inadequate, and the property of thermoplastics will become inadequate if [than the 200 weight sections] more. Moreover, stabilizers, such as various additives, for example, an anti-oxidant, an ultraviolet ray absorbent, light stabilizer, and a heat-resistant stabilizer, an antistatic agent, an anti-blocking agent, lubricant, a slipping agent, an antifogger, a foaming agent, a flameproofing agent, an inorganic bulking agent, an organic bulking agent, a color, a pigment, etc. can be blended with the resin constituent of this invention if needed.
[0056] It can manufacture to fitness by there being no limitation in the manufacturing method of the thermoplastics constituent of manufacturing method this invention, for example, kneading each component in the state of melting. As melting kneading equipment, a thing better known than the former, such as a roll mill of an open sand mold, a Banbury mixer of a non-open sand mold, an extruder, a kneader, and a continuous mixer, can be used.
[0057] Shaping of the thermoplastics constituent of fabricating method this invention can be performed with a conventional method. For example, extrusion molding, injection molding, compression molding, blow molding, rotational casting, or thermoforming from a sheet is performed using a single screw

extruder, a vent type extruder, 2 screw extruders, a two cone screw extruder, a ko kneader, PURATIFIKETA, mix TORUDA, a 2 shaft conical screw extruder, a planet **** extruder, a gearing die pressing appearance machine, a screwless extruder, etc. In these, injection molding is suitably employable especially. Thus, since the obtained mold goods, for example, a film, a sheet, molded product mold goods, etc. are excellent in shock resistance and a heat-sealing property, they can be used for various applications in the automobile field, the household-electric-appliances field, the film field, etc.

[0058]

[Example] Next, although an example and the example of a comparison show this invention concretely, this invention is not limited to the following example. First, in advance of manufacture of a thermoplastics constituent, the annular olefin system copolymer of the example of the following reference was manufactured.

[0059] Under the example nitrogen-gas-atmosphere mind of reference, in the room temperature, 15l. of toluene, triisobutylaluminum (TIBA) 23 millimol, zirconium tetrachloride 38 micromole, and tetrapod (pentafluorophenyl) boric acid anilinium 60 micromole were put into the 30l. autoclave at this sequence, and 2.4l. (considering as 2-norbornene 16 mols) of toluene solutions which contain 2-norbornene 70% of the weight continuously was added. The reaction was performed for 110 minutes, introducing ethylene continuously so that an ethylene partial pressure may become 8 kg/cm2, after carrying out a temperature up to 80 degrees C. After reaction termination, the polymer solution was thrown in in the 15l. methanol, and the polymer was deposited. This polymer was separated, it dried and the annular olefin system copolymer (a1) was obtained. The yield of an annular olefin system copolymer (a1) was 2.93kg. Polymerization activity was 845 kg/gZr.

[0060] The physical properties of the obtained annular olefin system copolymer (a1) were as follows. The norbornene content calculated from the ratio of the sum of the peak based on 5 and the methylene of the 6th place of the peak based on the ethylene which appears near 30 ppm of 13 C-NMR, and norbornene, and the peak based on the methylene group of the 7th place of the norbornene which appears near 32.5 ppm was 7.7-mol %. The degree of crystallinity which asked for the limiting viscosity [eta] measured in the 135-degree C decalin with 1.20 dl/g and an X-ray diffraction method was 1.0%. When the piece of measurement with a width of 4mm, a die length [of 40mm], and a thickness of 0.1mm was measured on the programming-rate a part for /and the frequency of 3.5Hz of 3 degrees C and it glass transition temperature (Tg) Asked for it from the peak of the loss modulus at this time (E"), using a Vibron 11made from Oriental ball DINGU-EA mold as a measuring device, Tg was 0 degree C. When weight average molecular weight Mw, number average molecular weight Mn, and molecular weight distribution (Mw/Mn) were searched for by polyethylene conversion at 1, 2, 4-trichlorobenzene solvent, and 135 degrees C, using Waters ALC / GPC150C as a measuring device, Mw was 69,600 and Mn was 35,900 and Mw/Mn=1.94. When the melting point (Tm) was measured in -50 degrees C - 150 degrees C with 10-degree-C programming rate for /by DSC of 7 by PerkinElmer, Inc. series, Tm was 72degree-C(broadcloth peak) **.

[0061] Next, the example and the example of a comparison using the annular olefin system copolymer a1 and other resin which were obtained in the above-mentioned example of reference are shown. The pellet of the resin which has the presentation shown in one to examples 1-6 and example of comparison 4 table 1 was manufactured. Next, using the obtained pellet, with the injection molding machine (ISby Toshiba Corp.25EP), injection molding was performed on condition that the laying temperature of 150 degrees C, the die temperature of 30 degrees C, and injection pressure (primary order [2nd]) 80 / 40 kg/cm2, and the test piece was obtained. The result of having measured physical properties, such as the tractive characteristics of the obtained test piece, a bending property, and an impact property, is shown in Table 1.

[0062] Here, measurement of each item was performed as follows.

Modulus of elasticity in tension: It measured based on JIS-K7113.

It measured based on flexural strength and bending elastic-modulus:JIS-K7200.

Eye ZODDO impact strength: It measured based on JIS-K7110.

Heat-sealing nature: The propriety of the welding when heat sealing for 10 seconds at the pressure of 1 kg/cm2 and predetermined temperature showed films with a thickness of 0.1mm.
[0063]
[Table 1]

	共重合体 a1 (重量%)	熱可塑性樹脂 (重量%)	エラストマー (重量%)	引張弾性率 (Kg/cm²)	曲げ強度 (Kg/cm²)	曲げ弾性率 (Kg/cm²)	アイゾッド衝撃強度 (Kg·cm/cm) 23℃	アイゾッド衝撃強度 (Kg·cm/cm) -40℃	ヒートシール温度 (℃)	ヒートシール性
実施例1	25	PP*1 75	0	11000	261	10070	53	3.9	156	可
実施例2	10	PP*1 75	*2 15	10680	249	9880	48	5.7	156	可
実施例3	5	PP*1 75	*3 20	10200	234	9420	46	6.7	156	可
実施例4	20	HDPE*4 80	0	11300	150	6000	*7 NB	4.9	132	可
実施例5	20	L-LDPE*5 80	0	6800	110	3400	NB	5.8	120	可
実施例6	20	PS *6 80	0	25500	310	20000	2.2	1.5	102	可
比較例1	0	PP*1 100	0	13100	330	12500	7.5	2.9	156	否
比較例2	0	HDPE*4 100	0	14800	200	8000	NB	3.8	132	否
比較例3	0	L-LDPE*5 100	0	9300	140	5000	NB	5.4	120	否
比較例4	0	PS *6 100	0	38100	400	32400	1.5	1.0	102	否

＊1ポリプロピレン ：出光石油化学社製 J－750H2 MI 10 (g/10分) (230℃, 2.16Kg)
＊2エチレン・プロピレン共重合体：日本合成ゴム社製 EP－961SP ムーニー粘度 (ML_{1+4} 100 ℃) 63
＊3EPDM ：日本合成ゴム社製 EP－57P ムーニー粘度 (ML_{1+4} 100 ℃) 88
＊4高密度ポリエチレン ：出光石油化学社製 440M MI 0.02 (g/10分) (190℃, 2.16Kg)
＊5直鎖低密度ポリエチレン ：出光石油化学社製 0368R MI 3.36 (g/10分) (190℃, 2.16Kg)
＊6ポリスチレン ：出光石油化学社製 HF・10 MI 3.87 (g/10分) (190℃, 2.16Kg)
＊7NB ：破壊せず

[0064]

[Effect of the Invention] As explained above, the thermoplastics constituent of this invention has the outstanding shock resistance and the outstanding heat-sealing property.

[Translation done.]